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PATENT SPECIFICATION

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NO DRAWINGS

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Int. Cl.:—C 08 g 21/04, C 08 g 53/10 // C 08 j

The inventors of this invention in the sense of being the devisers thereof within the meaning of Section 16 of the Patents Act 1949, are:— WULF VON BONIN, Schildgen, Katerbachstrasse 36, Germany, HELMUT PIECHOTA, Leverkusen, Opladener Strasse 75, Germany, both are German Citizens.

COMPLETE SPECIFICATION

Polyurethane Foams

We, FARBENFABRIKEN BAYER AKTIENGESELLSCHAFT, of Leverkusen-Bayerwerk, Germany, a body corporate organised under the Laws of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to polyurethane foams and to a process for their production.

The production of polyurethane foams has been hitherto proposed by the well-known polyisocyanate polyaddition process wherein an organic polyisocyanate is reacted with, for example, a polyhydric polyalkylene ether if desired in the presence of a blowing agent such as water; activators such as tertiary amines and/or organo metallic compounds; emulsifying agents and the like. Suitable processes for the preparation of polyurethane foams and the like may be found in *Angewandte Chemie* 59, 257 (1948) and in the pocketbook "Bayer-Kunststoffe", second edition, Page 25 (1959). It is known to produce both elastomeric and rigid foam plastics as well as various plastics between these two extremes.

The polyurethane foams based on polyisocyanates are advantageously prepared by mixing the liquid components which may be partially reacted in a first step to prepare a prepolymer or mixed substantially simultaneously in a single working step with a blowing agent to prepare a foamed plastic. The previously known foams have many advantageous properties but sometimes suffer from the disadvantage of relatively low breaking elongation, tensile strength and age resistance.

It is, therefore, an object of this invention to provide polyurethane foams which have improved physical properties.

Other objects of the invention include providing polyurethane foams having reduced inflammability as well as a method for the preparation of cellular polyurethane plastics in which polyhydric polyalkylene ethers are used.

According to the invention there are provided polyurethane foams prepared by a process which comprises reacting an organic polyisocyanate in the presence of a blowing agent with a modified polyhydric polyalkylene ether which has been prepared by effecting polymerisation of one or more ethylenically unsaturated monomers *in situ* in a polyhydric polyalkylene ether. The invention also provides a process for the preparation of polyurethane foams by a process in which an ethylenically unsaturated monomer is mixed with a polyhydric polyalkylene ether and then polymerized prior to bringing about reaction between the polyhydric polyalkylene ether and an organic polyisocyanate in the presence of a blowing agent. The modified polyhydric polyalkylene ether may be used either alone or mixed with additional polyhydric polyalkylene ether or other polyol. The ethylenically unsaturated polymerisable monomers having a molecular weight of up to about 500 are preferred.

The polymers produced *in situ* are preferable partially dissolved or suspended as

[Price

homopolymers in the polyhydric polyalkylene ethers which are preferably polyalkylene ether glycols. It is also possible to use graft copolymers such as those disclosed in the U.S. Patent 3,033,841, British Patent 874,130 and German Patents, 1,077,430; 1,105,179; 1,084,917 and 1,111,394. It could not be anticipated that such graft polymers and mixtures of ethylenically unsaturated monomers with polyhydric polyalkylene ethers would produce cellular polyurethane plastics having improved physical properties and particularly breaking elongation, tensile strength and resistance to aging.

Any suitable polyhydric polyalkylene ether having graft copolymers and/or mixtures of ethylenically unsaturated monomers dissolved in or bonded to the polyhydric polyalkylene ether may be used. Any suitable ethylenically unsaturated monomer containing one or more polymerisable double bonds may be used, for example, aromatic vinyl compounds such as styrene; olefinic hydrocarbons such as ethylene, propylene, 1-butylene, 2-hexene, 1,4-hexadiene, 1,3-butadiene, and 2-pentene; vinyl esters such as vinyl acetate, vinyl propionate and the like; vinyl halides such as vinyl chloride, vinylidene dichloride and the like; acrylic acid, methacrylic acid and derivatives thereof such as methacrylate, acrylate acrylonitrile; and monomers with more than one vinyl group, for example triallyl cyanurate, and glycol dimethyl acrylate. The ethylenically unsaturated monomers may be used either alone or mixed with one another.

Any suitable polyhydric polyalkylene ether may be used in admixture with these ethylenically unsaturated monomers but it is preferred to use substantially linear polyalkylene ether glycols which preferably have a molecular weight of from about 400 to about 5000 and an hydroxyl content of from about 0.5 to about 15 per cent. The polyhydric polyalkylene ethers formed by the condensation of alkylene oxides, for example, ethylene oxide, propylene oxide, 1,2- and 1,3-butylene oxide, styrene oxide, epichlorohydrin, and tetrahydrofuran. A polypropylene ether glycol having a molecular weight of from 500 to 2000 is particularly preferred. These alkylene oxides may be polymerised by the addition of an initiator in accordance with procedures well known in the art or they may be condensed with polyhydric alcohols or amines, for example, ethylene glycol, 1,2-propylene glycol, trimethylolpropane, 1,4-butane diol, 1,2,4-butane triol, glycerine, sorbitol, sucrose, glucose, alpha-methyl-D-glucoside, pentaerythritol, castor oil, ethanol amine, diethanol amine, triethanol amine, aniline, 2,4-tolylene diamine, 2,6-tolylene diamine, 4,4'-methylenediamine; alkylene diamines such as, for example, ethylene diamine, tetramethylene diamine, hexamethylene diamine and the like. Mixtures of the linear and branched polyhydric polyalkylene ethers of the various types may also be used. Moreover, it is possible to use prepolymers formed from the polyhydric polyalkylene ethers mentioned above which have been reacted with a less than equivalent amount of a reactive group to prepare compounds containing inter alia, urethane groups, and ester groups. Examples are the reaction product of toluylene diisocyanate with an excess of a low molecular weight polypropylene ether glycol or the reaction product thereof with a dicarboxylic acid such as adipic acid. A suitable toluylene diisocyanate for this purpose is a mixture of 80 per cent 2,4- and 20 per cent 2,6-toluylene diisocyanate and a suitable polypropylene ether glycol is one having a molecular weight of about 425.

The modification of the polyhydric polyalkylene ether is carried out in a straightforward manner by dissolving the ethylenically unsaturated monomer in the polyhydric polyalkylene ether preferably in an amount of 1 to 60 per cent by weight in particular from 1 to 30 per cent by weight and then bringing about polymerisation of the ethylenically unsaturated monomer while it is mixed with the polyhydric polyalkylene ether. It is desirable to carry out the polymerisation in the absence of oxygen but it is not necessary to carry out the reaction in the absence of air even though this, too, is desirable. Initiators for the polymerisation reaction include the usual radical formers such as peroxides, e.g. lauroyl peroxide, benzoyl peroxide, dicumyl peroxide and the like as well as nitrogen containing initiators such as azodiisobutyronitrile. Redox systems may also be used such as benzoyl peroxide-diethyl toluidine and polymerisation systems utilising metal ions as catalysts such as ferrous-ferric iron systems. Initiation of the polymerisation may also be effected by high energy radiation. The radical formers may be used in any convenient amount but it is preferred to use from 0.01 to 15 per cent by weight and preferably from 0.1 to 5 per cent by weight based on the weight of the polymerisable vinyl compound. Any order of addition may be used but it is convenient to dissolve the polymerisation initiator in the vinyl compound or in other words, the ethylenically unsaturated monomer, and then combine this mixture with the polyhydric polyalkylene ether which is then homogenised and polymerised by heating preferably in the absence of air. Occasionally, it may be desirable to filter off some polymer agglomerates which are formed in the reaction mixture before the polyhydric polyalkylene ether containing the polymerizate is reacted with an organic polyisocyanate.

As stated above, these polyhydric polyalkylene ethers containing the polymerisable may be used either alone or mixed with other organic compounds containing active hydrogen groups as determined by the Zerewitinoff method. Any suitable type of the latter may be used such as, for example, linear or branched polyesters including polyester amides, unmodified polyhydric polyalkylene ethers, reaction products of the amino alcohols, diamines, hydroxyamines or the like with alkylene oxides or carboxylic acids to prepare various mixtures of polyols. Particularly suitable polyesters are those formed from polyhydric alcohols such as ethylene glycol, 1,3-propylene glycol, 1,4-butane diol, glycerine, trimethylolpropane, 1,2,6-hexane-triol, pentaerythritol, sorbitol, and dipentaerythritol reacted with polycarboxylic acids such as adipic acid, sebacic acid, succinic acid, 1,4-benzene dicarboxylic acid, 1,3,5-benzene tricarboxylic acid and the like. Suitable polyester amides are prepared by reacting amino alcohols with the aforementioned polycarboxylic acids or by reaction thereof with mixtures of amines and polyhydric alcohols. Any suitable amino alcohol may be used such as ethanol amine, propanol amine and the like. Any suitable diamine may be used such as ethylene diamine, propylene diamine, toluylene diamines such as 2,4-toluylene diamine, hydroxyamines such as diethanol amine. Another way to make polyester amides is to include in the reaction mixture leading to the production of a polyester some amino carboxylic acid such as, for example, alanine and the like. The polyesters, polyester amides and the like may contain hetero atoms such as are obtained by including thiodiglycol in the reaction components, double or triple bonds such as are obtained by using ethylenically or acetylenically unsaturated compounds such as, for example, 3-hexene-1,6-diol and 3-hexyne-1,6-diol. In addition, saturated or unsaturated fatty acids and hydroxy acids may be included in the reaction mixture, for example, oleic acid, propionic acid, and the like as well as fatty alcohols, for example, lauryl alcohol.

The unmodified polyhydric polyalkylene ethers described above may also be used in admixture with the modified polyhydric polyalkylene ethers.

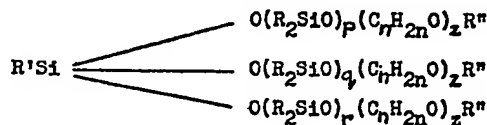
Suitable organic polyisocyanates, preferably organic diisocyanates for the preparation of the polyurethane plastics of the invention include, for example, ethylene diisocyanate, ethylidene diisocyanate, propylene-1,2-diisocyanate, cyclohexylene 1,2-diisocyanate, m-phenylene diisocyanate, 2,4-toluylene diisocyanate, 2,6-toluylene diisocyanate, 3,3'-dimethyl-4,4'-biphenylene diisocyanate, 3,3'-dimethoxy-4,4'-biphenylene diisocyanate, 3,3'-diphenyl-4,4'-biphenylene diisocyanate, 4,4'-biphenylene diisocyanate, 3,3'-dichloro-4,4'-biphenylene diisocyanate, p,p', p''-triphenylmethane triisocyanate, 1,5-naphthalene diisocyanate, furfurylidene diisocyanate or polyisocyanate in a blocked or masked form such as the bis-phenyl carbamates or 2,4- or 2,6-toluylene diisocyanate, p,p'-diphenylmethane diisocyanate, p-phenylene diisocyanate, and 1,5-naphthalene diisocyanate. It is preferred to use aromatic diisocyanates and the commercially available mixture of toluylene diisocyanates which contain 80 per cent 2,4-toluylene diisocyanate and 20 per cent 2,6-toluylene diisocyanate or 4,4'-diphenylmethane diisocyanate is most preferred.

Crude isocyanates may be used such as, for example, crude toluylene diisocyanate obtained by the phosgenation of a mixture of toluylene diamines or crude 4,4'-diphenyl methane isocyanates (also referred to as crude polyphenyl methane diisocyanate) obtained by the phosgenation of crude 4,4' diphenyl methane diamine. Crude 4,4'-diphenyl methane diamine is the reaction product of aniline and formaldehyde in the presence of HCl and contains some tri- and higher polyamines. A preferred unrefined or crude isocyanate is one having from 26 to 33 per cent free —NCO and an amine equivalent of 120 to 150, such as, for example, a product having 32 per cent free —NCO and an amine equivalent of 140. A specified crude isocyanate suitable for use in accordance with the present invention may be obtained by reacting 60 parts of aniline with 25 parts of formaldehyde (37 per cent aqueous) and 74 parts of HCl (30 per cent aqueous) at a temperature of 90°C to 100°C for 1.5 to 2 hours and then reacting this product with NaOH and separating out the crude amine. 100 parts of phosgene are then reacted with the crude amine until a product having an amine equivalent of 135 and containing 31 per cent free —NCO is obtained. The free excess phosgene and substantially all of the solvents used, if any, are then removed. When toluylene diisocyanates, for example, are produced by conventional phosgenation of the corresponding diamines, a product containing 90 per cent 2,4- and 2,6-toluylene diisocyanate and the balance a crude residue of imidazoles and the like resulting in the phosgenation is obtained from the phosgenator. This product may also be used. It is preferred to phosgenate a mixture of ortho- and para-toluylene diamines. A specific product is the undistilled reaction mixture obtained by the phosgenation of 80 per cent 2,4- and 20 per cent 2,6-toluylene diamine containing 90 per cent of a mixture of 80

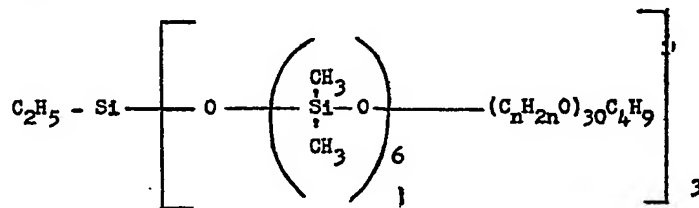
per cent 2,4- and 20 per cent 2,6-toluylene diisocyanate and the balance polymers incapable of accurate analysis.

The commercially available crude 4,4'-diphenyl methane diisocyanate (also referred to as polyphenyl methane polyisocyanate) having an assay of about 90 per cent maximum, an amine equivalent of 141 maximum, 0.04 to 0.4 per cent by weight hydrolysable chloride, 0.1 to 0.6 per cent by weight total chloride and having a flash point of 430°F. may be used. As pointed out above, when toluylene diisocyanate, for example, is produced by conventional phosgenation of the corresponding diamine, a product containing about 90 per cent 2,4- and 2,6-toluylene diisocyanate is obtained from the phosgenator. Of course, the product from the phosgenator is subjected to distillation to remove the solvent so that a product having 90 per cent 2,4- and 2,6-toluylene diisocyanate is obtained. The initial product from the phosgenator in most cases contains 80 per cent by weight of solvent. The 2,4- and 2,6-toluylene diisocyanate may be mixed with any suitable amount of the residue obtained if the isocyanate is refined and then reconstituted. In this event, it is preferred to have at least 50 per cent of the refined isocyanate present. The crude polyisocyanates disclosed in U.S. Patent 2,950,307 are suitable.

The preparation of the polyurethane foams of the invention is carried out in the manner well known in the art by a rapid and thorough mixing of the organic polyisocyanate with the polyhydric polyalkylene ether containing polymerisates in the presence of water or other blowing agents to prepare a polyurethane foam. It is preferred in the production of the polyurethane foams to include an activator such as a tertiary amine or an organo metallic compound. Any suitable tertiary amine may be used such as, for example, dimethyl benzyl amine, N-methyl-N'(N-dimethyl amino ethyl) piperazine, triethylene diamine, permethylated diethylene triamine and the like. Any suitable organo metallic compound may be used such as those disclosed in U.S. Patent 2,846,408. It is preferred to use organo tin compounds such as, for example, dibutyl tin dilaurate, dibutyl tin di-2-ethyl hexoate, stannous octoate, stannous oleate and the like. It is preferred to use a foam stabiliser for the production of the cellular polyurethane plastics such as, for example, sulphonated castor oils and sodium salts thereof. Where polyhydric polyalkylene ethers are included in the reaction mixture to prepare a cellulose polyurethane foam, it is preferred to employ a silicone oil such as that disclosed in U.S. Patent 2,834,748 within the scope of the formula



wherein R, R' and R'' are alkyl radicals having 1 to 4 carbon atoms; p, q and r each have a value of from 4 to 8 and $(C_nH_{2n}O)_z$ is a mixed polyoxyethylene oxypropylene group containing from 15 to 19 oxyethylene units and from 11 to 15 oxypropylene units with z equal to about 26 to 34. Most preferred is a compound having the formula

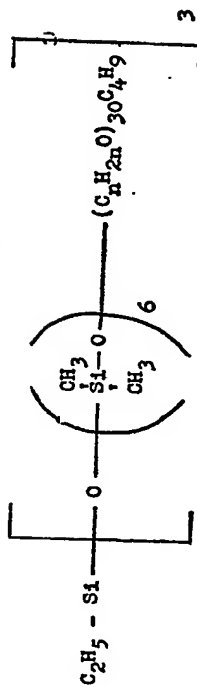


wherein $(C_nH_{2n}O)$ is a mixed polyoxyethylene and oxypropylene block copolymer containing about 17 oxyethylene units and about 13 oxypropylene units.

The cellular polyurethanes of the invention are useful for the production of both sound and thermal insulation, cushions such as automobile seat topper pads and the like. The most important advantage is the noticeable improvement in the mechanical properties of the cellular polyurethane plastics. The mechanical properties which are most improved are tensile strength, elongation, compression strength, impact elasticity and residual compressive deformation. These properties are improved independent of the density of the cellular polyurethane plastic and are due solely to this modification.

The improvement in the physical properties is demonstrated by reference to Table

1. A cellular polyurethane plastic was prepared by reacting 200 parts of a polypropylene ether glycol having a molecular weight of 200 and an hydroxyl number of 61 with 77 parts of a mixture of 80 per cent 2,4- and 20 per cent 2,6-tolylene diisocyanate, 6 parts of water, 0.5 part of endoethylene piperazine, 0.2 part of dibutyl tin dilaurate and 2 parts of a silicone having the formula



wherein $(\text{C}_4\text{H}_9\text{O})$ is a mixed polyoxyethylene and oxypropylene block copolymer containing about 17 oxyethylene units and about 13 oxypropylene units. These components were mixed together on a machine mixer as disclosed in U.K. Patent Specification No. 769,682 and the resulting mixture was allowed to foam and cure. In addition to a control which contained no ethylenically unsaturated monomer in the polypropylene ether glycol, the polypropylene ether glycol was modified with 7 per cent by weight of the ethylenically unsaturated monomer shown in the table. All parts in the foregoing formulation are by weight. After cooling, the properties were measured on the sample and are shown in the table (the foams are hand samples).

TABLE I

	Unmodified	Vinyl Chloride	Vinyl Acetate	Acrylonitrile	Methyl Methacrylate	Styrene
Density kg/cm ³	32	31	33	34	31	33
Tensile strength kg/cc	1.0	1.1	1.1	1.1	0.9	1.2
Breaking elongation per cent	260	415	330	330	250	365
Impact elasticity	31	32	34	35	34	—
Compression strength 20% 40% 60% compression	16—25—39	21—24—38	26—31—43	17—27—42	18—23—34	23—28—41
Residual compressive deformation	26	23	13	41	29	—

Another advantage of the polyalkylene ether glycols is that these foamed plastics have a higher resistance to organic solvents and are therefore suitable for coating fabrics because in addition to their good resistance to hydrolysis, they are also resistant to organic solvents used in dry cleaning processes. Previously only cellular polyurethanes formed from polyesters could be used for this purpose. The polyester based cellular polyurethanes are indeed resistant to organic solvents but they have poor resistance to hydrolysis. In Table 2 below, the modification of the behaviour of cellular polyurethanes toward solvents is demonstrated where the polyurethane plastic is based on the modified polyalkylene ether glycol of the invention. In the following Table, (a) is a control sample prepared by the same procedure as the sample of Table 1 but based on 200 parts of a mixture of 50 per cent by weight of the polypropylene ether glycol having a molecular weight of 2000 and an hydroxyl number of 61 and 50 per cent by weight of a trihydric polyalkylene ether prepared by condensing propylene oxide with trimethylolpropane, said trihydric polyalkylene ether having an hydroxyl number of 56 and a molecular weight of 3000, 85 parts of 80 per cent 2,4- and 20 per cent 2,6-toluylene diisocyanate, 0.5 part of endoethylene piperazine, 0.2 part dibutyl tin dilaurate, 4 parts of the silicone oil used in the Example for Table I and 6 parts of water. The test specimen for Table 2 is 13.4 × 4.5 × 0.7 cm. Sample (b) was prepared by the same procedure and the same components only 15 per cent by weight of acrylonitrile having been polymerised in situ in the polyether mixture.

TABLE 2

	(a)	(b) Acrylonitrile
Content (% by weight)	0	15
Change in % after 24-hour storage in:		
trichloro ethylene	42.0	29.0
perchloro ethylene	25.0	17.0
benzene	11.0	8.2
gasoline	35.0	25.0

The cellular polyurethanes of the invention have in addition other properties which are substantially and often strikingly improved by utilisation of polyols of the invention which had polymerised therein or therewith an ethylenically unsaturated monomer. The chlorine or other halogen containing compounds impart reduced flammability to both flexible and rigid cellular polyurethanes; often to a degree that they will not support combustion. Further, the cellular polyurethanes have better heat sealing characteristics as evidenced by greater ease of uniformly heat sealing as well as more tenacious bonds to itself and other materials such as fabrics, plastics such as nylon, and the like. This latter property makes the cellular polyurethanes all the more useful for desirable commercial applications including the flame lamination process of U.S. Patent 2,957,793. In order that the invention may be further understood, the following examples, in which parts are given by weight unless otherwise indicated, are given by way of illustration only:—

35 Production of the starting materials 35

(a) 2000 parts of a linear propylene glycol ether (molecular weight about 2000, —OH number about 61.2), 1000 parts vinyl chloride, $\text{CH}_2 = \text{CHCl}$, and 2.0 parts benzoyl peroxide were placed in a mixing autoclave. Stirring was continued for 6 hours at 75°C, and followed by evacuation in order to remove residual monomeric vinyl chloride. The reaction product was a milky substance, having the following analysis: 40

—OH number, 56.6
Cl content, 3.8%
Vinyl polymer content, 10%.

- (b) 1000 parts of a linear polypropylene glycol ether (molecular weight about 2000, —OH number about 61.2) and 3.0 parts benzoyl peroxide, dissolved in 100 parts vinyl acetate, were placed in a stirring tube under a nitrogen atmosphere. Stirring was continued for 21 hours at 75°C and then nitrogen was blown through the clear solution for one hour, in order to remove unreacted vinyl acetate. The reaction product had an —OH number of 56.6. 5 5
- (c) The process used was the same as under (b), but a solution of 3.0 parts azidiisobutyronitrile in 100 parts vinyl acetate was used. The product had an —OH number of 56.6. 10
- (d) Process as under (b), but acrylonitrile was used instead of vinyl acetate. The —OH number of the product was 48.7. 10
- (e) Process as under (d), but lauroyl peroxide was used instead of benzoyl peroxide, and the polymerisation was effected at 60°C. The product had an —OH number of 48.6. 15
- (f) Process as under (b), but methyl methacrylate was used instead of vinyl acetate. The product had an —OH number of 56.3. 15
- (g) Process as under (b), but styrene was used instead of vinyl acetate. After polymerisation was terminated any polymerisation agglomerates which separate were removed by filtering. The product had an —OH number of 52.7. 20
- (h) Process as under (f), but 200 parts methyl methacrylate were used, and in addition to 3.0 parts benzoyl peroxide 0.2 parts of N,N'-dimethyl-p-toluidine as well. The product had an —OH number of about 55.0. 20
- (i) Process as under (a), but a branched polypropylene glycol ether (molecular weight about 2500—3000, —OH number about 56.8) was used as starting material. The end product had an —OH number of 49.1. 25 25
- (k) Process as under (d), but the starting product was a branched polypropylene glycol ether (molecular weight 4000, —OH number 47.3). The product had an —OH number of 37.7. 30
- (l) Process as under (b), with a mixture of 60 parts vinyl acetate and 40 parts acrylonitrile replacing the vinyl acetate. The product had an —OH number of 52.6. 30

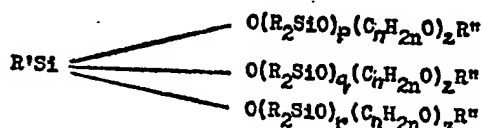
EXAMPLES 1 to 11

- 200 parts of modified polyalkylene glycol ether as specified under (a) to (l) above were reacted with stirring with 0.5 parts endoethylene piperazine, 0.2 part dibutyl tin dilaurate, 2.0 part polysiloxane-polyalkylene glycol ester, 6.0 part water, and the amounts of toluylene diisocyanate given in Table 3. The reaction took place after thorough mixing. The product was poured into a mould and foamed plastic was formed having the properties given in Table 3. 35 35

Example	Amount of toluylene diisocyanate	Starting material	Volumetric weight kg/m ³	tensile strength kg/cc	Elongation	Compression strength 20% 40% 60%	Intact elasticity %	Residual compressive deformation %
1	78	a	31	1.1	415	21 24 38	32	23
2	78	b	33	1.1	330	26 31 43	34	13
3	78	c	32	1.1	325	27 30 41	38	10
4	75	d	34	1.1	330	17 27 42	35	41
5	75	e	31	1.2	330	25 28 41	37	10
6	78	f	31	1.1	370	20 24 35	34	19
7	76	g	33	1.2	365	23 28 41	36	12
8	77	h	33	1.0	265	24 29 43	34	41
9	75	i	33	0.8	165	33 36 48	36	8
10	72	k	34	1.0	140	29 42 61	41	15
11	76	l	31	1.2	330	26 29 41	38	14

WHAT WE CLAIM IS:—

1. A process for the production of a polyurethane foam which comprises reacting an organic polyisocyanate in the presence of a blowing agent, with a modified polyhydric polyalkylene ether prepared by effecting polymerisation of one or more ethylenically unsaturated monomers in situ in a polyhydric polyalkylene ether.
2. A process as claimed in claim 1 in which ethylenically unsaturated monomers are employed having a molecular weight of not greater than 500.
3. A process as claimed in claim 1 or claim 2 in which the polyhydric polyalkylene ether is substantially linear and has a molecular weight of 400 to 5000 and an hydroxyl content of 0.5 to 15 per cent by weight.
4. A process as claimed in claim 3 in which the polyhydric ether is a polypropylene ether glycol having a molecular weight of from 500 to 2000.
5. A process as claimed in claim 1 or claim 2 in which the unsaturated monomers are styrene, methyl methacrylate, vinyl acetate, vinyl chloride or acrylonitrile.
6. A process as claimed in any one of claims 1 to 5 in which the polymerisation of the ethylenically unsaturated monomers in situ in the polyhydric polyalkylene ether is initiated in the presence of 0.01 to 15% by weight based on the weight of the ethylenically unsaturated monomers of a radical forming compound.
7. A process as claimed in claim 6 in which the radical forming compound is an organic peroxide.
8. A process as claimed in claim 6 in which the radical forming compound is azodiisobutyronitrile.
9. A process as claimed in claim 6 in which a redox system is used to initiate the polymerisation.
10. A process as claimed in any one of claims 1 to 5 in which the polymerisation of the ethylenically unsaturated monomers in situ in the polyhydric polyalkylene ether is initiated by means of high energy radiation.
11. A process as claimed in any one of claims 1 to 10 in which the organic polyisocyanate is a mixture of 80% 2,4- and 20% 2,6-toluylene diisocyanate.
12. A process as claimed in any one of claims 1 to 10 in which the organic polyisocyanate is a polyphenyl methane polyisocyanate.
13. A process as claimed in any one of claims 1 to 10 in which the organic polyisocyanate is a crude mixture of polyphenyl methane polyisocyanates obtained by the phosgenation of crude 4,4'-diphenyl methane diamine.
14. A process as claimed in any of claims 1 to 13 in which the polyurethane-forming reaction is effected in the presence of a catalyst.
15. A process as claimed in claim 14 in which the activator is a tertiary amine.
16. A process as claimed in claim 14 in which the activator is an organo tin compound.
17. A process as claimed in any one of claims 1 to 16 in which a foam stabiliser is used.
18. A process as claimed in claim 17 in which the foam stabiliser is a sulphonated castor oil or a salt thereof.
19. A process as claimed in claim 17 in which the foam stabiliser is a silicone oil of the general formula



wherein R, R' and R'' are alkyl radicals having 1 to 4 carbon atoms, p, q and r each have a value of from 4 to 8 and $(C_nH_{2n}O)_z$ is a mixed polyoxyethylene oxypropylene group containing from 15 to 19 oxyethylene units and from 11 to 15 oxypropylene units with z equal to from 26 to 34.

20. A process as claimed in claim 19 in which the silicone oil stabiliser has the formula